Multilayer Electrodes with Metalized Polymer Current Collector for High-Energy Lithium-Ion Batteries with Extreme-Fast-Charging Capability

Oak Ridge National Laboratory | National Transportation Research Center

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2022 DOE Annual Merit Review, Project ID = BAT551

Timeline
- Start: September 3, 2021
- End: September 3, 2024
- Percent Complete: 15%

Budget from AMO
- Total project funding: $3000k
- FY22 - $1000K

Barriers
- Reduce battery pack cost to <$100/kWh
- Reduce charging time to <15 min

RELEVANCE

- Reduce battery pack cost by lowering the cost of current collectors
- Increase battery gravimetric energy density by reducing the weight of current collectors
- Develop multi-layer electrodes to improve mass transport for fast charging

OBJECTIVES

Main Objective: Develop a multilayer graphite anode and NMC622 cathode coated on thin metalized polymer films for extreme fast charging (XFC) application
- Cell energy density >230 Wh/kg and >300 Wh/kg under XFC and 0.2C, respectively
- Less than 20% capacity fade in 500 cycles under XFC conditions
- 7% cost reduction on battery pack

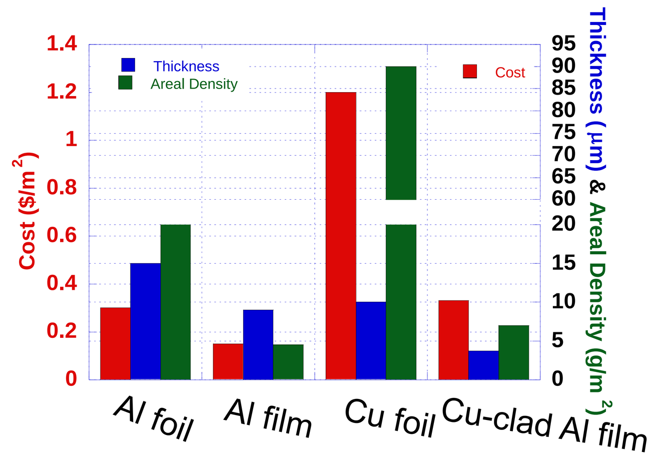
PROJECT MILESTONES

Status	SMART Milestones	Description
12/31/21	Milestone 1.1 Completed	Create a cost model of metalized film production.
3/31/22	Milestone 1.2 Completed	Design two-layer anode based on numerical simulations to explore various configurations (layer thickness, porosity).
6/30/22	Milestone 1.3 On track	Fabricate > 100 m with 250 mm width metalized PET film.
6/30/22	Milestone 1.4 On track	Assemble and characterize >20 Gen 1 pouch cells.
9/30/22	Go/No-Go Decision On track	Gen 1 Pouch Cell: >160 Wh/kg and <20% capacity fade in 500 XFC cycles.

TECHNICAL APPROACH

Problems:
- State-of-the-art chemistries are capable of XFC but cell energy density is limited (e.g. < 180 Wh/kg) due to limitation on mass transport.
- Low areal loading in the cells for XFC application increases cell cost.

Technical approach and strategy:
- Develop and fabricate metalized polymer films as current collectors
  - 91% reduction in weight
  - 49% reduction in volume
  - 68% reduction in cost



- Improve mass transport with multiple electrolyte architecture

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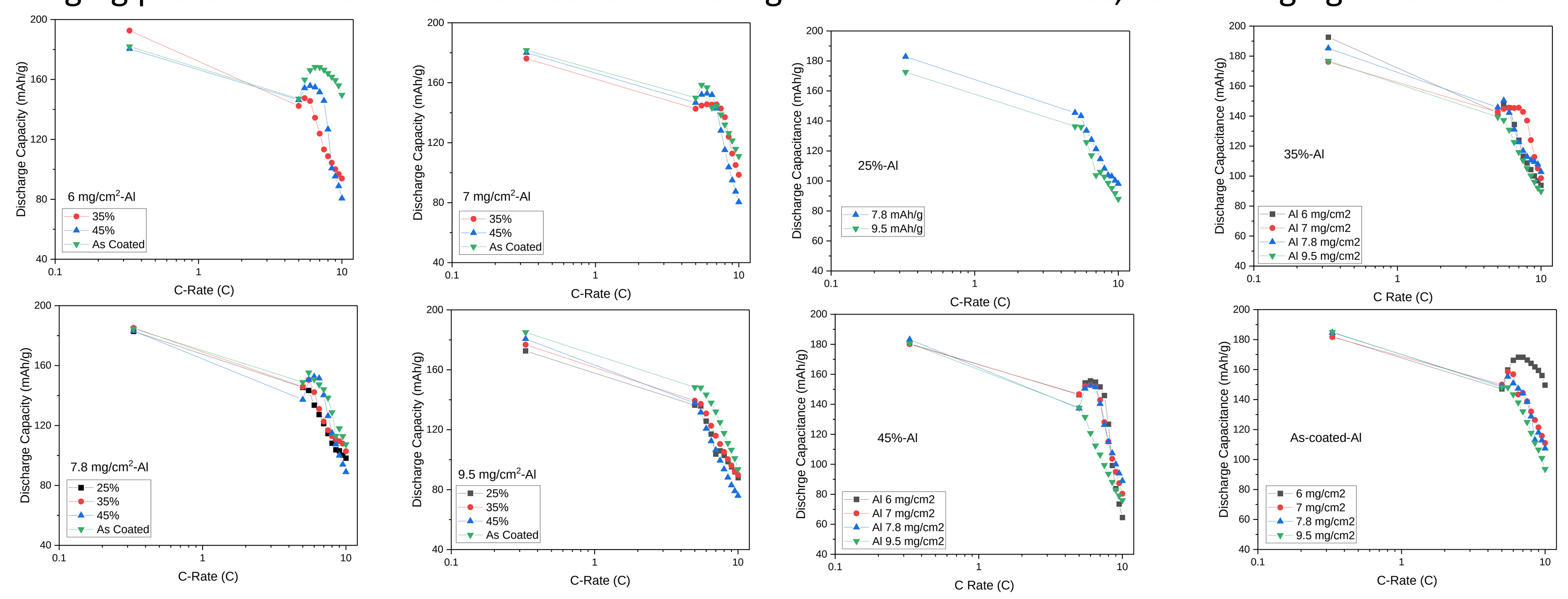
TECHNICAL ACCOMPLISHMENTS

Executive Summary:
- A cost model for metalized films projecting $0.22/m² and $0.32 m² for metalized aluminum and copper films, respectively. Compared to the conventional aluminum ($0.3/m²) and copper foils ($1.2 m²), it enables 63.3% cost reduction in current collector.
- A roll of aluminum metallized film (305 mm x 500 m) has been fabricated.
- A 3D model has been developed to simulate lithium distribution.
- Both anode and cathode with four areal loadings on metal foils and metalized films, respectively.
- Initial electrochemical test shows 17.9% improvement in maximum gravimetric energy density when replacing the Al foil with Al-PET.

Fabrication of Baseline Cathodes and Evaluation of Their Performance Under Extreme Fast Charging

- Cathode composition: NMC622/carbon black/PVDF: 90/5/5 wt
- XFC charging protocol: constant current constant voltage with various C-rates, total charging time of 10 min

Specific Capacity

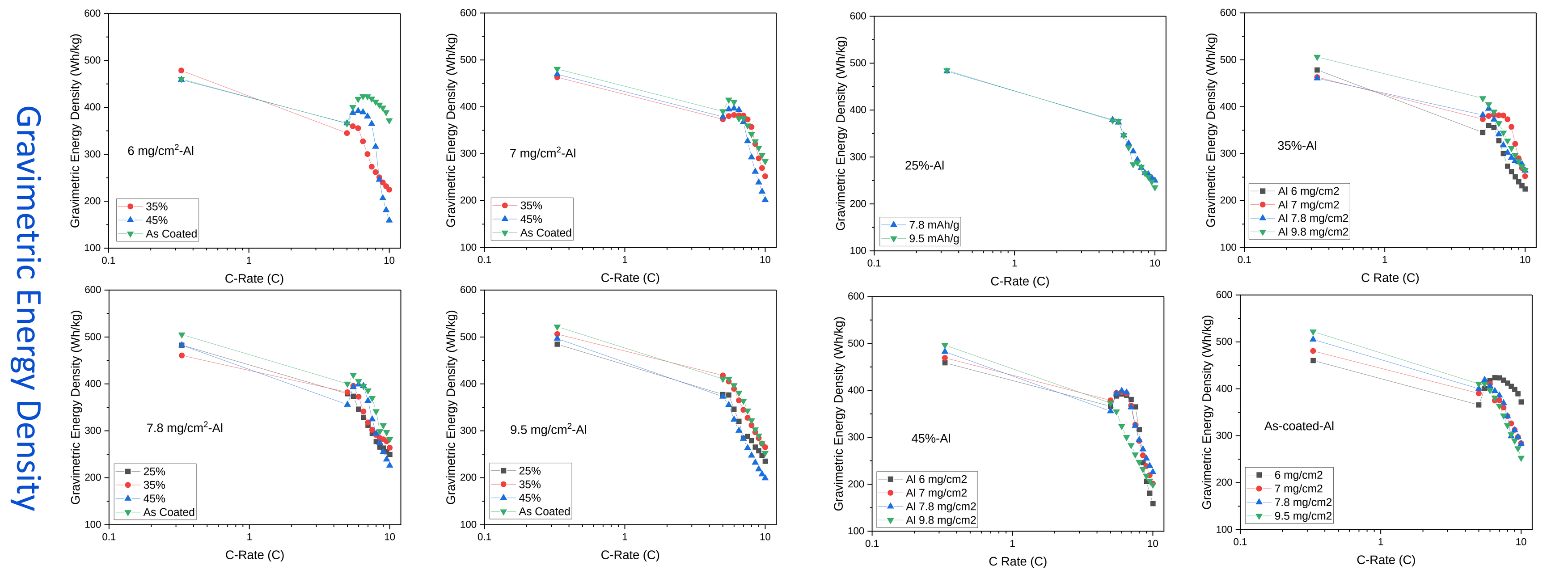


Porosity effect—Higher porosity benefits capacity under XFC

Mass loading effect—Higher capacity with lower mass loading under XFC

Gravimetric Energy Density

Normalized to the total mass of cathode coating and half of the Al foil

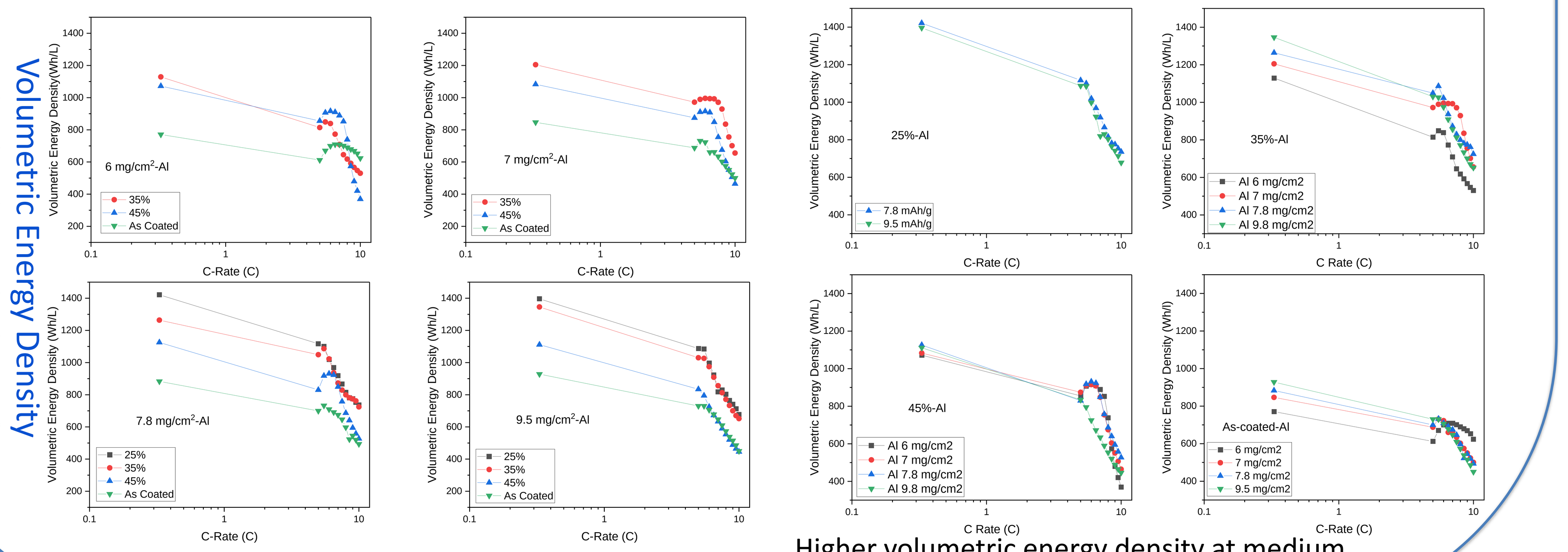


Porosity effect—In general, higher porosity favors gravimetric energy density under XFC

Mass loading effect—Higher gravimetric energy density with higher areal loading at low C-rate, however, there is no clear trend under XFC

Volumetric Energy Density

Normalized to the total volume of cathode coating and half of the Al foil



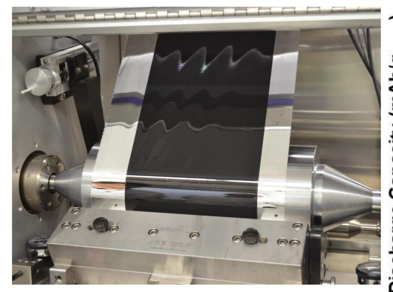
Higher volumetric energy density with lower porosity

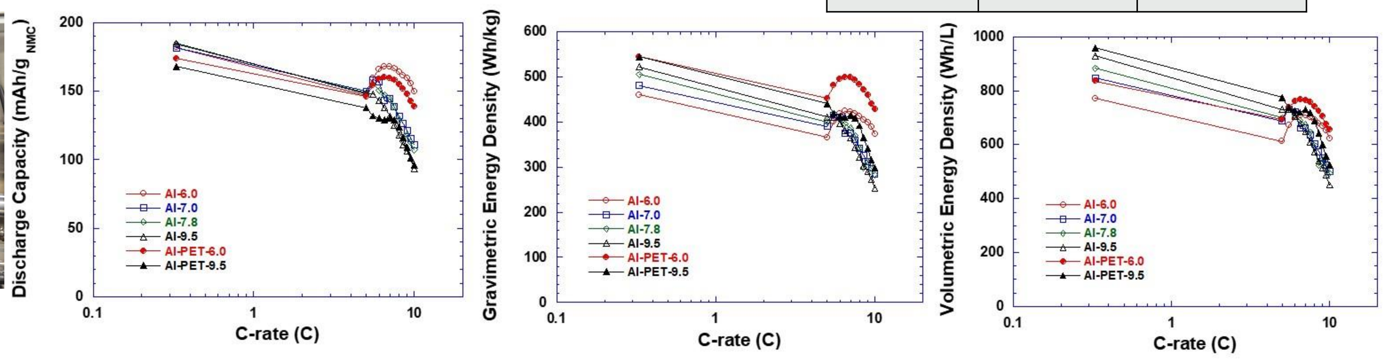
Higher volumetric energy density at medium porosity and areal loadings (7.8 mg/cm² & 35%)

Successful Coating NMC622 Cathode on Metalized Polymer

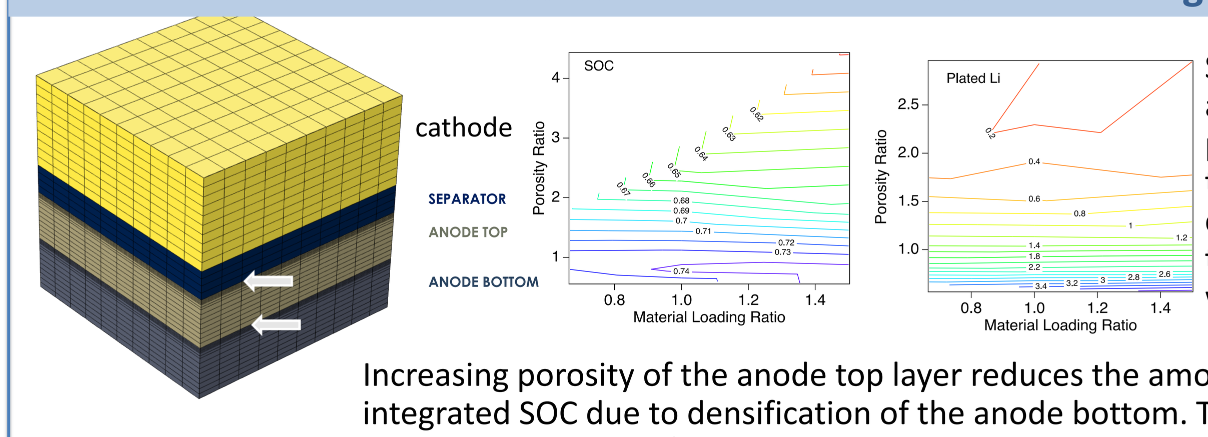
Replacing Al foil with Al-PET film enables 17.9% and 5.6% improvement in maximum gravimetric and volumetric energy density under XFC, respectively

Substrate	Thickness	Areal Mass
Al foil	15 um	4.175 mg/cm²
Al mPET	8 um	1.128 mg/cm²





Prediction of Electrode Utilization and Lithium Plating via Numerical Simulation



Series of numerical simulations to investigate average SOC in anode and total thickness of plated lithium at the end of 3C charge. The total anode loading and thickness were kept constant, but the ratio of porosities, $\varepsilon_2/\varepsilon_1$ and the ratio of loadings in anode layers, g_2/g_1 were varied.

Increasing porosity of the anode top layer reduces the amount of plated lithium, but it also reduces the integrated SOC due to densification of the anode bottom. The optimal arrangement seems with porosity ratio of 35%/33% and material loading fraction of 40%/60% in [anode top/anode bottom].

SUMMARY

- Baseline NMC622 and graphite electrodes have been fabricated on Al and Cu foils.
- NMC622 and graphite electrodes have been fabricated on metalized polymer films.
- Effect of mass loading and porosity on XFC performance has been evaluated on NMC622 cathodes.
- Initial results demonstrated 17.9% and 5.6% improvement in maximum gravimetric and volumetric energy density under XFC, respectively, when replacing Al foil with metalized polymer film.

FUTURE WORK

Any proposed future work is subject to change based on funding levels

Remainder of FY22
- Complete testing baseline electrodes with various areal loadings and porosities under XFC
- Down-select the areal loading with highest energy densities
- Fabricate two-layer electrode and assemble Gen 1 cells
- Evaluate the electrochemical performance of the Gen 1 cells

Into FY23
- Develop a numeric model to simulate the two-layer electrodes with two passes (16 combinations)
- Develop metalized films on new polymers
- Fabricate two-layer electrodes via two passes and assemble Gen 2 cells
- Evaluate the electrochemical performance of the Gen 2 cells

COLLABORATIONS

Oak Ridge National Laboratory | Soteria

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